ON THE SIMPLE TENSILE DEFORMATION OF AN INCOMPRESSIBLE RUBBER MATRIX FILLED WITH NON-ADHERENT RIGID SPHERES OF UNIFORM SIZE DISTRIBUTION

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INTRODUCTION

Two striking features revealed in a photograph (cf Figure 1) of a thin film of rubber binder highly filled with glass beads are: a) that the growth of voids around particles increases with increasing strain and b) that the preferred direction of the void growth seems to be in the direction of the applied macroscopic strain. It is obvious that the local stress field around particles in a deformed composite is not as high as it would be if the binder did not pull away from the filler particles. On the other hand, because of the high rigidity of the particles relative to the binder, the local stress field in the binder will still be significantly higher than the average macroscopic stress field. It is of interest to define both this stress field and the associated dilatation in terms of a simple model.

In pursuing this point, it seemed reasonable to the authors to assume that the filler particles are infinitely rigid, that the rubber matrix is incompressible, and, that, since the voids line up with the direction of pull in simple tension, the local stress field is essentially simple tensile. While this latter assumption is not as rigorous an approximation as the first two assumptions, it is still not without an excellent basis. That is as follows. For a Mooney-Rivlin material, which is a reasonable constitutive form for any type of binder that is currently used in propellant technology, the form of the true stress deviator for a homogeneous stress field, is given by:

$$\bar{\sigma}_i - \bar{\sigma}_j = G\left[f\,(\lambda_i^2 - \lambda_j^2) + (1-f)\left(\frac{1}{\lambda_i^2} - \frac{1}{\lambda_j^2}\right)\right] \tag{1}$$

In the case of uniaxial deformation, $\lambda_j^2 = 1/\lambda_i$ (2)

In the case of strip-biaxial deformation (pure shear), $\lambda_j^2 = 1/\lambda_i^2$ (3)

In either case as λ_i becomes large, the true stress deviator becomes proportional to λ_i^2; and thus, the two stress fields cannot be distinguished in this limit.

Proceeding from these assumptions, a model is developed which predicts the type of dilatation-stretch behavior one should expect for a rubber matrix loaded with non-adherent rigid filler particles. Since there is a tendency for filler particles in a real composite propellant to dewet in a fashion as Farris has shown (Ref. 1), the model must be modified to include the effect of non-uniform dewetting and the way in which this can be done is described. A comparison of the prediction of the model with data obtained by Farris is offered. In addition, the strain energy of a composite space deformed under the above assumptions is calculated and the associated simple stress-deformation relation is shown. A comparison of the prediction of this relation with available data will be presented in a later publication.

GEOMETRY OF THE MODEL

Consider a finite composite space which is filled with a mass of incompressible rubbery matrix M_r, in which are uniformly distributed rigid spherical particles of diameter a, and of total mass M_f. If the densities of these two components are respectively ρ_r and ρ_f, then the volume of this space is given by:

$$V = \frac{M_r}{\rho_r} + \frac{M_f}{\rho_f} \tag{4}$$

The number of particles in this volume is given by:

$$n = \frac{M_f/\rho_f}{\frac{\pi}{6} a^3} \tag{5}$$

so that the average volume of space occupied by one filler particle and its fair share of surrounding rubber matrix is given by:

$$\frac{V}{n} = \frac{\pi}{6} a^3 \left(1 + \frac{\rho_f}{\rho_r} \frac{M_r}{M_f}\right) \tag{6}$$

The fractional volume occupied by filler particles is given by:

$$\nu_f = \frac{1}{1 + \frac{\rho_f}{\rho_r} \frac{M_r}{M_f}} \tag{7}$$

so that Equation (6) may be rewritten

$$\frac{V}{n} = \frac{\pi a^3}{6 \nu_f} \tag{8}$$

Now consider in this composite space a unit cell in the shape of a rhombohedron (Ref. 2). This cell is a generator for a face-centered cubic lattice and is readily obtained by surrounding a central lattice point with twelve neighbors, - six around, three above, and three below, - at each of the twelve adjacent lattice points. The volume enclosed by these twelve lattice ports includes the entire sphere at the center plus a fraction of each of the twelve neighbor spheres plus the interstitial rubber. This volume may be readily calculated by observing that the unit cell may be partitioned into six tetrahedra and six square-based pentahedra. If the lattice distance be R, then the volume of each tetrahedron is $\sqrt{2}\, R^3/12$, while the volume of each pentahedron is $\sqrt{2}\, R^3/6$. Thus the total volume of the rhombohedral cell is $(3/\sqrt{2})\, R^3$.

Now the fraction of each neighbor sphere enclosed within the unit cell is readily calculated by observing that a homeomorphism of the unit cell is a face-centered cube, which contains exactly four spheres (Ref. 3), the combined volume of which is $(2/3)\, \pi\, a^3$. On the other hand the volume of the cube is $2\sqrt{2}\; R^3$, so that the volume fraction of loading is given by:

$$\nu_f = \frac{\pi}{3\sqrt{2}} \left(\frac{a}{R} \right)^3 \tag{9}$$

or

$$\left(\frac{R}{a} \right)^3 = \frac{\pi}{3\sqrt{2}\;\; \nu_f} \tag{10}$$

We note from Equation (9) that, at the point of tangency, or when a = R, the close-packed volume fraction is $\pi/3\sqrt{2}$ or 74 %. The number of particles (p) in the cell is given by the product of the volume of the cell and the volume fraction of filler divided by the volume of a particle, or:

$$p = 3 = 1 + 12k \tag{11}$$

where k is the fraction of a particle entrained within the cell. This is readily seen to be 1/6. An alternative consistency statement is provided by noting that the number of particles in the cell (3) multiplied by the average volume per particle, given by (8) must be equal to the volume of the cell, and indeed this is so. In a real material, of course, the cells are not uniform in size, and the particles are not uniformly distributed, so that the parameter p may be treated as just that, namely a fitting parameter for the representation of data.

Now in order to provide a convenient framework for describing the deformation of such a composite space, it is useful to replace the rhombohedral cell by an equivalent spherical cell, of the same volume. This immediately defines the radius of the sphere (using Equation (10)) to be:

$$\left(\frac{R_S}{a} \right)^3 = \frac{p}{8\,\nu_f} \tag{12}$$

where we have taken the value of the rhombohedral cell to be $p/\sqrt{2}\; R^3$!

Within this sphere, we assume as before that one particle is entered at the center of the sphere, and that the other (p-1) particles are now uniformly distributed to form a shell within the outer radius of the sphere. The thickness of this shell is given by:

$$\frac{2t}{a} = \left(\frac{p}{\nu_f}\right)^{\frac{1}{3}} - \left(\frac{p}{\nu_f} - p + 1\right)^{\frac{1}{3}} \tag{13}$$

We shall now describe the contour of the cell after the application of a simple tensile deformation.

GEOMETRY OF THE DEFORMED CELL

We assume first that each cell comprising the entire bulk volume of the composite space deforms affinely, which means that the deformation of the outer boundary of each cell exactly parallels the deformation of the outer boundary of the entire bulk volume. Thus, if a simple tensile slab of propellant is stretched by a factor λ in the z-direction along its principal length, and the lateral dimensions are allowed to contract freely resulting in a volume ratio J_3, then each point within the bulk is assumed to move according to the mapping:

$$\begin{aligned} x' &= R' \sin\theta' \cos\varphi' = \sqrt{(J_3/\lambda)}\, R_S \sin\theta \cos\varphi = \sqrt{(J_3/\lambda)}\; x \\ y' &= R' \sin\theta' \sin\varphi' = \sqrt{(J_3/\lambda)}\, R_S \sin\theta \sin\varphi = \sqrt{(J_3/\lambda)}\; y \\ z' &= R' \cos\theta' = \lambda R_S \cos\theta = \lambda z \end{aligned} \tag{14}$$

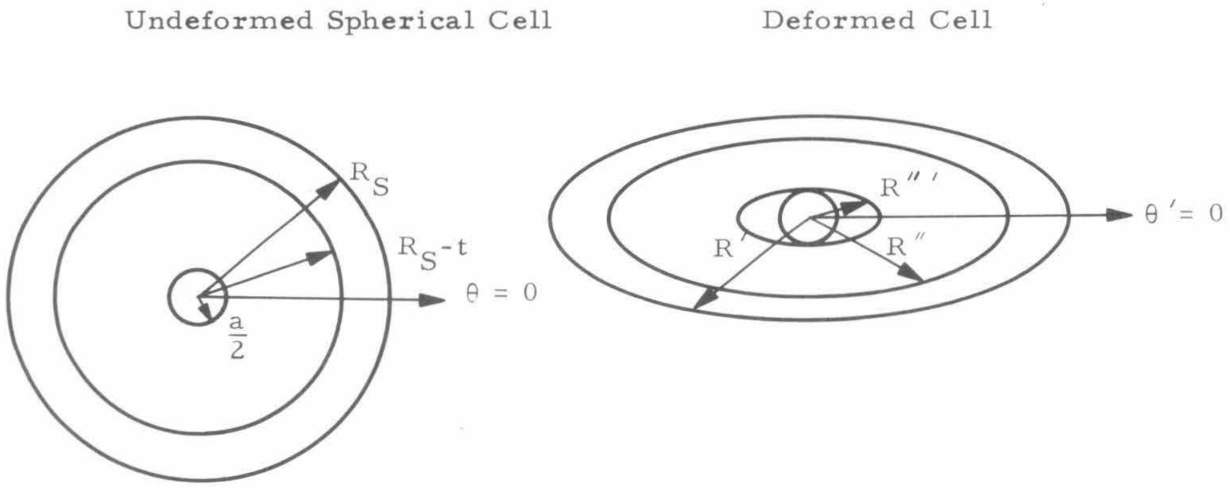
The transformation of the spherical coordinates is given by:

$$\begin{aligned} R' &= R_S \sqrt{\lambda^2 \cos^2\theta + (J_3/\lambda)\sin^2\theta} \\ \theta' &= \arctan \sqrt{(J_3/\lambda^3)}\tan\theta \\ \varphi' &= \varphi \end{aligned} \tag{15}$$

or inversely:

$$\begin{aligned} R_S &= R' \sqrt{(\cos^2\theta'/\lambda^2) + (\lambda/J_3)\sin^2\theta'} \\ \theta &= \arctan \sqrt{\lambda^3/J_3}\, \tan\theta' \\ \varphi &= \varphi' \end{aligned} \tag{16}$$

Along with this affine deformation of the outer boundary of the originally spherical cell, there will be a non-affine deformation of the space within the cell. The following sketch shows how the rubber pulls away from the central filler particle opening up a non-spherical void bounded by a surface described by the Radius R'''. Likewise the radius vector to the inner surface of the shell of neighbor particles is denoted by R''. Each of these radius vectors are functions of θ', which function we shall now determine.



By definition, the total volume of the deformed cell is J_3 times the volume of the undeformed cell. This ratio is measurable in the form of dilatation data and provides a very useful characterization of propellant materials. Analytically, this statement may be phrased locally as:

$$\pi\,(R'\sin\theta')^2\, d\,(R'\cos\theta') = J_3\,\pi\,(R_S\sin\theta)^2\, d\,(R_S\cos\theta) \qquad (17)$$

It is immediately verified that Equation (15) satisfies Equation (17) identically, which it must since J_3 is just the Jacobian of the transformation Equation (15). The solution to Equation (17) may also be written:

$$\left(\frac{R'}{a}\right)^3 = \frac{p}{8\,\nu_f}\left(\lambda^2\cos^2\theta + (J_3/\lambda)\sin^2\theta\right)^{3/2} \qquad (18)$$

Since the particles are incompressible, the volume of the shell must be conserved, so that

$$\pi\,(R'\sin\theta')^2 d\,(R'\cos\theta') - \pi\,(R''\sin\theta')^2 d(R''\cos\theta') = \pi\left[R_S^{\,3} - (R_S - t)^3\right]\sin^2\theta\; d(\cos\theta) \qquad (19)$$

Using Equation (12) and Equation (13), the solution to Equation (19) is given by:

$$\left(\frac{R''}{a}\right)^3 = \frac{1}{8}\left(\frac{p}{\nu_f} - \frac{p-1}{J_3}\right)\left(\lambda^2\cos^2\theta + \frac{J_3}{\lambda}\sin^2\theta\right)^{3/2} \qquad (20)$$

The rubber matrix is also incompressible, which statement leads to the relation:

$$\pi(R'' \sin\theta')^2\, d(R''\cos\theta') - \pi\,(R'''\sin\theta')^2\, d\,(R'''\cos\theta') = \pi\left[(R_S - \theta)^3 - (\tfrac{a}{2})^3\right]\sin^2\theta\, d(\cos\theta) \tag{21}$$

The solution to Equation (21) is given by:

$$\left(\frac{R'''}{a}\right)^3 = \frac{1}{8}\left[\frac{p}{\nu_f} + \frac{1}{J_3} - \frac{p}{\nu_f J_3}\right]\left(\lambda^2\cos^2\theta + \frac{J_3}{\lambda}\sin^2\theta\right)^{3/2} \tag{22}$$

Finally, we must demand that the volume of the void is related to the volume of the undeformed cell by the relation:

$$\pi(R'''\sin\theta')^2\, d(R'''\cos\theta') - \pi\,(\tfrac{a}{2})^3\sin^2\theta\, d\,(\cos\theta) = (J_3 - 1)\,\pi R_S^{\,3}\sin^2\theta\, d\,(\cos\theta) \tag{23}$$

which, in view of Equation (22), is an identity, thus establishing the self-consistency of all the previous relations.

Now according to the sketch above, $R''' = a/2$ at $\theta' = \pi/2$. Thus, Equation (22) leads immediately to the important relation:

$$\lambda^3 = J_3\left[1 + \frac{p}{\nu_f}\,(J_3 - 1)\right]^2 \tag{24}$$

This is the dilatation-stretch relation predicted by our simple model. Since the dilatation does not usually exceed ten percent, Equation (24) may be inverted with

$$J_3 = 1 + \vartheta \tag{25}$$

to yield:

$$J_3 - 1 \equiv \vartheta = \frac{-\left(\frac{1}{2} + \frac{p}{\nu_f}\right) + \sqrt{\left(\frac{1}{2} + \frac{p}{\nu_f}\right)^2 + (\lambda^3 - 1)\left(\frac{2p}{\nu_f} + \frac{p^2}{\nu_f^{\,2}}\right)}}{\frac{2p}{\nu_f} + \frac{p^2}{\nu_f^{\,2}}} \approx \frac{\lambda^3 - 1}{1 + \frac{2p}{\nu_f}} \tag{26}$$

The limiting slope of Equation (24) is obtained when $J_3 > 10$ (never achieved in practice), and is equal to:

$$\frac{d\,J_3}{d\lambda} \to \left(\frac{\nu_f}{p}\right)^{2/3} \tag{27}$$

Inspection of the behavior of Equation (24) up to large values of J_3 reveals that it is remarkably linear, and a fortiori even more so within the range of a few percent dilatation. Because of this linearity the limiting slope is a good approximation to the limiting slope evinced by propellant data. We predict a (2/3)-power dependance on filler concentration as opposed to

Farris' first power. The disparity is not marked.

In order to test the usefulness of Equation (24), data from Farris (Ref. 4) for propellants ANP-2969 and ANP-2874 loaded respectively with $\nu_f = 0.50$ and $\nu_f = 0.70$ were plotted on Figure 2. On this same figure the straight black lines represent the behavior of the function Equation (24) for $\nu_f/p = 0.50$ and $\nu_f/p = 0.70$ respectively. Apparently this indicates that the best fit is obtained with p = unity, which indicates that the model does not account for the dilatation of the neighboring particles. This could be done at the expense of a considerable increase in algebraic complexity. We choose to throw this binder onto the fitting parameter p, even though it loses some physical significance!

In fitting Farris' data, it was necessary to shift the origin of the actual data to the left by an arbitrary amount. This is to account for the delayed onset of dewetting inherent in propellant behavior. An improvement in this model in the direction of accounting for distributed dewetting could be provided by making p a function of λ. In line with Farris' suggestion, this function should be given by (cf Blatz (Ref. 3)):

$$p = \frac{p_{max}}{s\sqrt{2}} \int_{1-\overline{\lambda}/s\sqrt{2}}^{1-\overline{\lambda}/s\sqrt{2}} e^{-u^2} du \tag{28}$$

When Equation (28) is introduced into Equation (24) it becomes transcental. On the other hand, Equation (28) can be also introduced into Equation (26) resulting in an explicit form for dilatation as a function of stretch. Calculations are now proceeding along this line, based on the values of <u>s</u> given by Farris, and using p_{max} as the one and <u>only</u> fitting parameter. In view of the moderately strain approximation also displayed in Equation (26), it is possible to test rapidly the applicability of Equation (28) by plotting

$$\frac{\nu_f}{2} \left(\frac{\lambda^3 - 1}{\vartheta} - 1\right) \text{ vs } (\lambda - 1) \tag{29}$$

on probability paper. A linear relation can be obtained. Thus, the combination of our model with Farris' distribution function results in an excellent model for predicting real propellant data.

THE STRAIN ENERGY FUNCTION

Referring back to the sketch of the deformed cell, it is seen that the rubber in this cell is stretched by the factor:

$$\lambda_r \equiv \frac{R'' - R'''}{R_S - t - \frac{a}{2}} = \frac{\left(\frac{p}{\nu_f} - \frac{p-1}{J_3}\right)^{\frac{1}{3}} - \left[\frac{1}{J_3} + \frac{p\,(J_3 - 1)}{\nu_f J_3}\right]^{\frac{1}{3}}}{\left(\frac{p}{\nu_f} - p + 1\right)^{\frac{1}{3}} - 1} \times \tag{30}$$

$$\sqrt{\lambda^2 \cos^2\theta + J_3/\lambda \ \sin^2\theta} \equiv \sqrt{F} \cdot \sqrt{\lambda^2 \cos^2\theta + J_3/\lambda \ \sin^2\theta}$$

The average square stretch ratio is obtained as follows:

$$\langle \lambda_r{}^2 \rangle = \theta \int_{\pi/2}^{0} \lambda_r{}^2 \, d\,(\cos\theta) = \frac{F}{3}\,(\lambda^2 + \frac{2J_3}{\lambda}) \tag{31}$$

Thus the first stretch invariant is given by:

$$I_{1r} = (\lambda^2 + \frac{2J_3}{\lambda})\,F \tag{32}$$

where

$$F \equiv \left\{ \frac{\left(\frac{p}{\nu_f} - \frac{p-1}{J_3}\right)^{\frac{1}{3}} - \left[\frac{1}{J_3} + \frac{p(J_3-1)}{\nu_f J_3}\right]^{\frac{1}{3}}}{\left(\frac{p}{\nu_f} - p + 1\right)^{\frac{1}{3}} - 1} \right\}^2 \tag{33}$$

Likewise the second stretch invariant is given by:

$$I_{2r} = 3 \int_{\pi/2}^{0} \frac{d\,(\cos\theta)}{\lambda_r{}^2} = \frac{3}{F}\,\frac{\lambda}{\sqrt{J_3(\lambda^3 - J_3)}}\,\arctan\sqrt{\frac{\lambda^3 - J_3}{J_3}} \tag{34}$$

Assuming the rubber to be Mooney-Rivlin, the strain energy of the rubber is given by:

$$W_r = \frac{G}{2}\left[f\,(I_{1r} - 3) + (\,1-f)\,(\,I_{2r} - 3)\,\right] \tag{35}$$

And, since the strain energy function W, by definition, is based on unit volume of undeformed material, the strain energy, based on unit volume of the undeformed composite, is given by:

$$W = \frac{G}{2\,(1-\nu_f)}\left[f\,(I_{1r} - 3) + (\,1-f)\,(\,I_{2r} - 3)\,\right] \tag{36}$$

where f is a parameter, the significance of which (Ref. 5) is given by noting that:

$$\frac{G}{2} f \equiv C_1 \text{ , the first Mooney-Rivlin parameter}$$

$$\frac{G}{2} (1-f) = C_2\text{, the second Mooney-Rivlin parameter}$$

Empirically, it is observed that f is negative for highly filled materials.

The stress-stretch relationsfor simple tension are obtained as follows. In W, J_3 is replaced wherever it appears, by $\lambda \lambda_e^2$. Then we have:

$$\sigma = \frac{\partial W}{\partial \lambda} \tag{37}$$

$$O = \frac{\partial W}{\partial \lambda_e} \tag{38}$$

Equation (37) defines the tensile stress. And, Equation (38) provides another form of dilatation-stretch relation. This latter relation should evince essentially the same behaviour as that of Equation (24). It is actually only as good as the assumption made in representing the simple tensile stretch field by the averages taken in Equations (31) and (34). The calculated prediction of Equation (37) and Equation (38) will be presented in a subsequent report.

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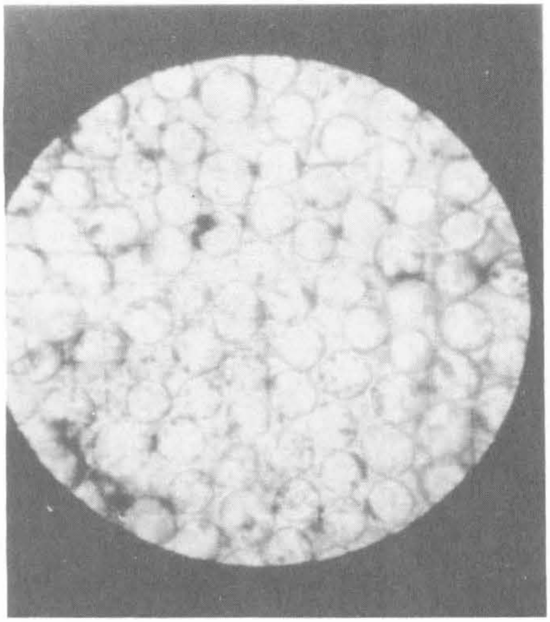
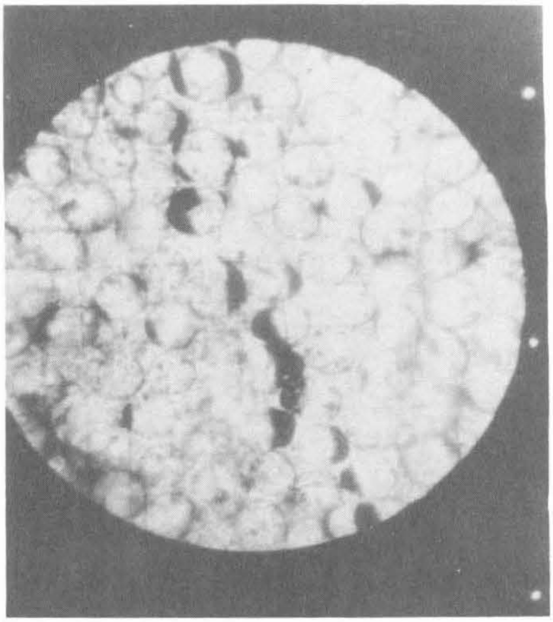
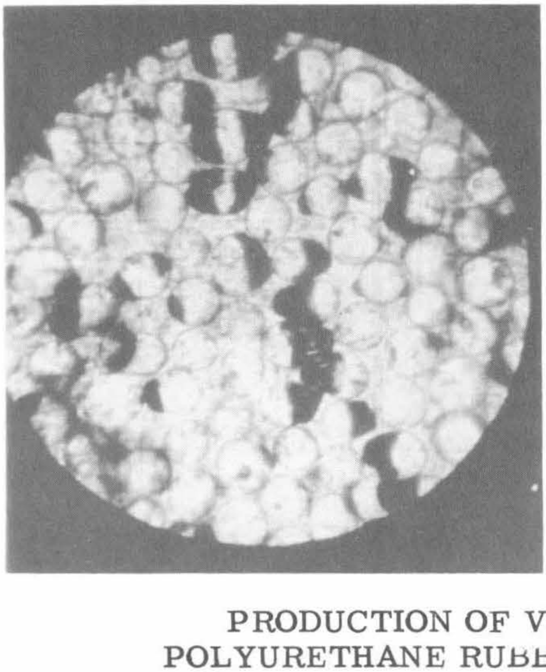
DIRECTION OF STRAIN

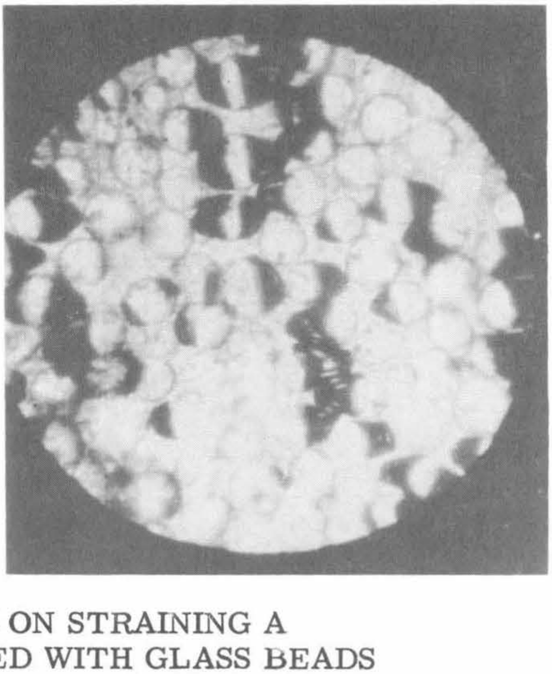




5% ELONGATION

10% ELONGATION





PRODUCTION OF VACUOLES ON STRAINING A
POLYURETHANE RUBBER FILLED WITH GLASS BEADS

15% ELONGATION

25% ELONGATION

FIGURE 1.

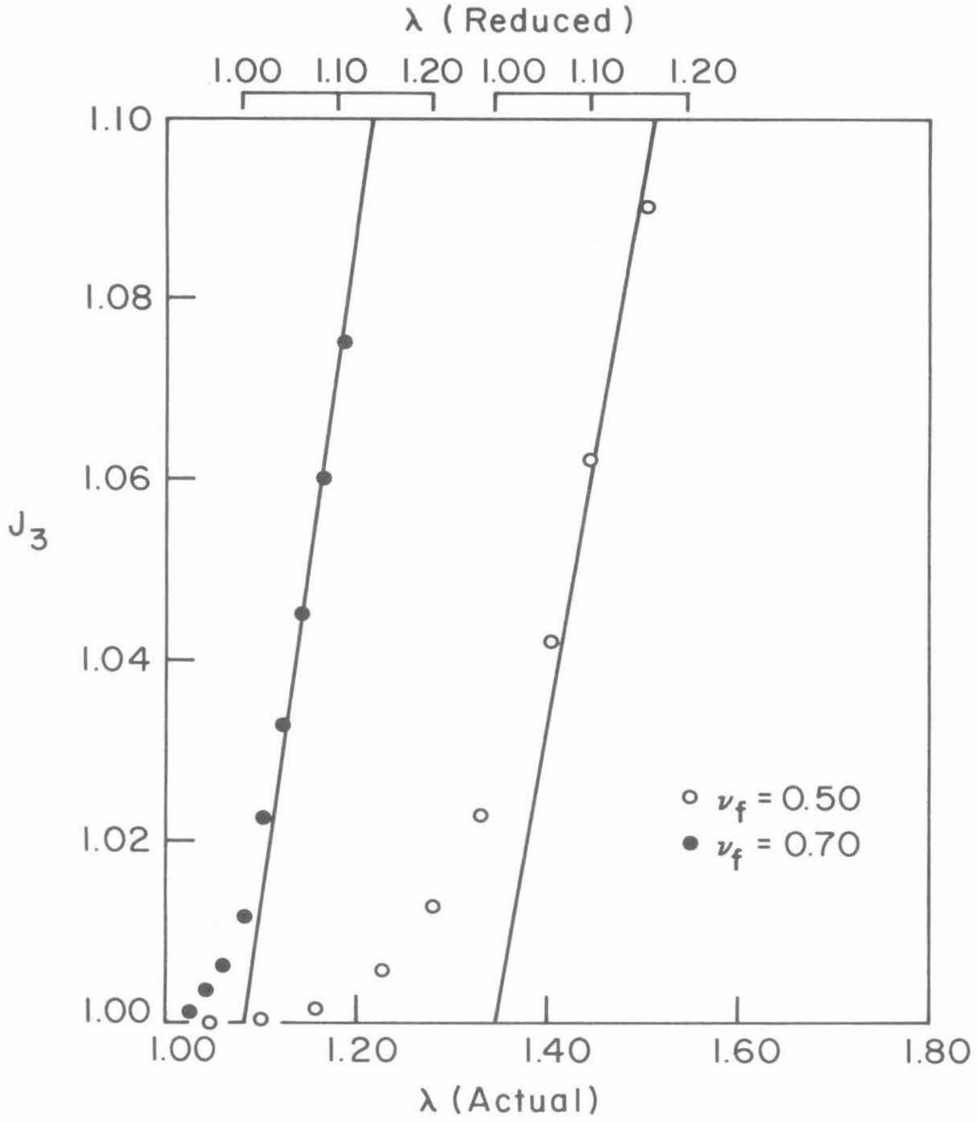


FIGURE 2. VOLUME RATIO PLOTTED AS A FUNCTION OF STRETCH RATIO. THE EXPERIMENTAL POINTS OF FARRIS (REF. 4) REFERS TO λ (ACTUAL). THE THEORETICAL CURVES (EQUATION (24)) WERE SHIFTED ("REDUCED λ") BY ARBITRARY AMOUNTS TO ACCOUNT FOR DELAYED DEWETTING OF REAL PROPELLANTS.

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Unclassified complete papers accepted for, and available prior to, the fifth meeting of the ICRPG Working Group on Mechanical Behavior (of solid propellants) are presented. The papers relate to four broad areas: (1) relationship between microstructure and propellant mechanical properties, (2) new or improved experimental methods for characterization, (3) failure criteria, and (4) structural analysis.

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